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Conversion of Solid Waste to Fuels

by

C. B. Benham
and
J. Diebold
Propulsion Development Department

JANUARY 1976

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FOREWORD

This report describes a solid waste research project conducted during the period FY73 through FY74 by the Naval Weapons Center (NWC), China Lake, California. The effort was sponsored by the Defense Advanced Research Projects Agency (DARPA) under ARPA Order No. 2772.

This report has been reviewed for technical accuracy by J. D. Andrews.

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(U) *Conversion of Solid Waste to Fuels*, by C. B. Benham and J. Diebold. China Lake, Calif., NWC, January 1976, 30 pp. (NWC TP 5797, publication UNCLASSIFIED.)

(U) Economic and practical processes for recovering energy from solid waste were studied. Two promising fuels were identified – polymer gasoline and methanol. A nominal 10-pound-per-hour pyrolysis system was constructed and tested. Preliminary cost analyses and studies of the effects of population and energy market value on fuel costs were also conducted.

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SUMMARY

As part of the Naval Weapons Center's Total Energy Community (TEC) Program, the solid waste conversion effort involved exploring processes for recovering energy from solid waste which would be practical and economical on military bases. Emphasis was placed on conversion of solid waste and development of liquid fuels for mobile power plant use.

Two promising fuels were identified -- polymer gasoline and methanol. Preliminary process flow sheets for these fuels were developed. These indicated that an energy conversion efficiency of 66% for polymer gasoline and 58% for methanol could be achieved.

Preliminary cost analyses were made for conversion facilities consisting of a front-end system for removal of metals and glass, a pyrolysis system for production of synthesis gas and a synthesis module for conversion to fuel. Effects of population and energy market value on fuel costs were studied.

A nominal 10-pound-per-hour pyrolysis system was constructed and put into operation. Checkout runs were made using a shredded-paper feed at feed rates to 10 pounds/hour.

INTRODUCTION AND BACKGROUND

The recovery of energy from solid waste is attractive because it addresses the problems of both waste disposal and energy shortages. Energy recovery is particularly attractive for remote military bases where transportation costs add significantly to the total fuel cost. Energy recovery from solid waste has been exploited in its simplest form for several years by the Naval Station, Norfolk, Va. There, trash is burned in a water-walled incinerator to produce steam.

Incineration of solid waste for steam production is a viable option for some military bases, and will become attractive to more bases as old oil and gas fired boilers are retired from service. Package boilers designed for solid waste combustion are becoming available and should be considered in modernization plans for steam-generating plants on military bases.

However, this particular study was aimed at conversion of solid waste to a higher-grade fuel, i.e., gasoline or a suitable substitute. The objectives of this work were to (1) identify candidate fuels obtainable from solid waste, (2) determine the processes required for conversion to the candidate fuels, (3) establish the realistic yields and efficiencies of the processes, and (4) assess and compare the overall economics of the conversion processes.

SOLID WASTE CHARACTERIZATION

Before it can be determined what fuels can be derived from solid waste, assessment of the composition of solid waste must be made. The composition varies from one geographical area to another and depends on the time of year. For the purposes of this study, a composition was established which was felt to be representative of the data available at this time. This "average" composition is listed in Table 1.

A large fraction of the organic solid waste is paper and cardboard which is chiefly cellulose with an empirical formula of $C_6H_{10}O_5$. Ultimate analyses on solid waste, made by the Bureau of Mines,[1] yielded an empirical formula of $C_6H_{9.1}O_{3.1}N_{0.13}S_{0.014}$. Thus, a ton of the dry organic part of solid waste contains 1,082 pounds carbon, 136 pounds hydrogen, 748 pounds oxygen, 27 pounds nitrogen, and 7 pounds sulfur.

The quantity of solid waste generated on a per capita basis also varies both seasonally and geographically. For this study, an average generation rate of 5.3 pounds per person per day was assumed. While these figures correspond to estimates for the local Indian Wells Valley in Kern County, California area (61 tons/day for a population of 23,000), they fall within ranges of generation rates quoted in the literature for other parts of the country.[2] A study for any particular location should begin with the determination of generation rates and composition of solid waste for that specific area.

ENERGY IMPACT

The energy contained in solid waste represents 2-3% of the total energy budget in the United States.[3] The potential impact of using *all* of the solid waste generated by NWC and the outlying civilian population is shown in Table 2.

---

[1]Sanner, W. S., et.al. "Conversion of Municipal and Industrial Refuse Into Useful Materials by Pyrolysis", RI-7428, U.S. Dept. of the Interior, Bureau of Mines (1970).

[2]George, Patricia C., "Solid Waste America's Negleeted Pollutant", Nation's Cities, June 1970.

[3]Anderson, Larry L., "Energy Potential from Organic Wastes: A Review of the Quantities and Sourees", IC-8549, U.S. Dept. of the Interior, Bureau of Mines (1972).

TABLE 1. Characterization of Solid Waste

Constituent	Weight, %	Value, $/ton
Moisture	27.0	. . .
Ferrous	5.5	40
Aluminum	0.5	300
Non-ferrous	0.2	120
Glass	6.6	7
Organics	60.2	. . .

NOTE: Total revenue from inorganic fraction is $4.40/ton for raw refuse. Energy content of raw solid waste is 9 MBtu/ton.

TABLE 2. Potential Impact of Solid Waste as an Energy Source at NWC.

Energy use at NWC (1972)	Energy quantity, MBtu[a]	Energy in solid waste,[b] MBtu[a]	Energy fraction available from solid waste[b]
Interruptible natural gas and No. 6 fuel oil	511,000	192,600	0.38
Firm natural gas and propane	487,000		0.40
Gasoline and diesel (military only)	127,000		1.52
Electricity	1,239,000[c]		0.16
Total	2,364,000		0.08

[a]Million British thermal units.

[b]Based on 4,500 Btu/lb of raw solid waste and 100% conversion of energy in solid waste.

[c]33% conversion efficiency from KW_t to KW_e.

CANDIDATE FUELS

A brief review of any standard chemical engineering handbook reveals that, in addition to gasoline, the fuels that should be considered for mobile power plants are the alcohols and perhaps benzene and some of its derivatives. It is instructive to compare the two classes of fuels derivable from solid waste, namely hydrocarbons and alcohols. Figure 1 is a plot of volumetric energy content versus molecular weight for the alkanes, alcohols, and some aromatic compounds. The same comparison on a weight basis is presented in Figure 2. The boiling point range of these compounds is shown in Figure 3, along with the boiling range for gasoline.

The use of methyl and ethyl alcohol as gasoline blending stocks is discussed in a report by the American Petroleum Institute.[4] Table 3, based partly on this report, is a list of several properties for these alcohols plus iso-octane and benzene. The main arguments against the use of alcohols are based on unfavorable economics, but the recent rapid escalation of gasoline prices may invalidate this argument. Benzene is a high value fuel because it can be used to increase the octane rating of gasoline when tetraethyl lead usage is reduced to meet pollution standards. Purified methanol and benzene also have a considerable value as petrochemical feedstocks. An upper limit on the amount of each fuel type that can be obtained from solid waste can be established based on the composition of the organic material. This information is listed in Table 4 for three different constraints placed upon the conversion process. If yields are based on the hydrogen content of the solid waste, the amounts obtainable for the four fuels are as shown in the first row. The carbon-limit values are based on using the water-gas shift reaction to produce additional hydrogen from excess carbon. However, if no outside energy source is available, the maximum yields are determined by energy limitations, as shown in the bottom row. From these rudimentary mass and energy considerations alone, one can obtain upper limits on the amount of fuels obtainable from solid waste. However, other practical constraints in the process flowsheet reduce the achievable yields well below the theoretical maximum. These practical considerations are discussed in a later section of this report.

GENERAL CONCEPTS IN CONVERSION

Thus far, estimates of quantities of fuels based on elementary mass and energy considerations have been made. The practical problem of how to effect the conversion from a solid material, which is primarily cellulose, to either a liquid hydrocarbon or an alcohol fuel remains to be discussed. It is helpful at this point to categorize available methods for extracting and converting the organic material into more useful forms.

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[4]Keller, J G., et.al, "Use of Alcohol in Motor Gasoline – A Review", American Petroleum Institute. Publication No. 4082 (August 1971).

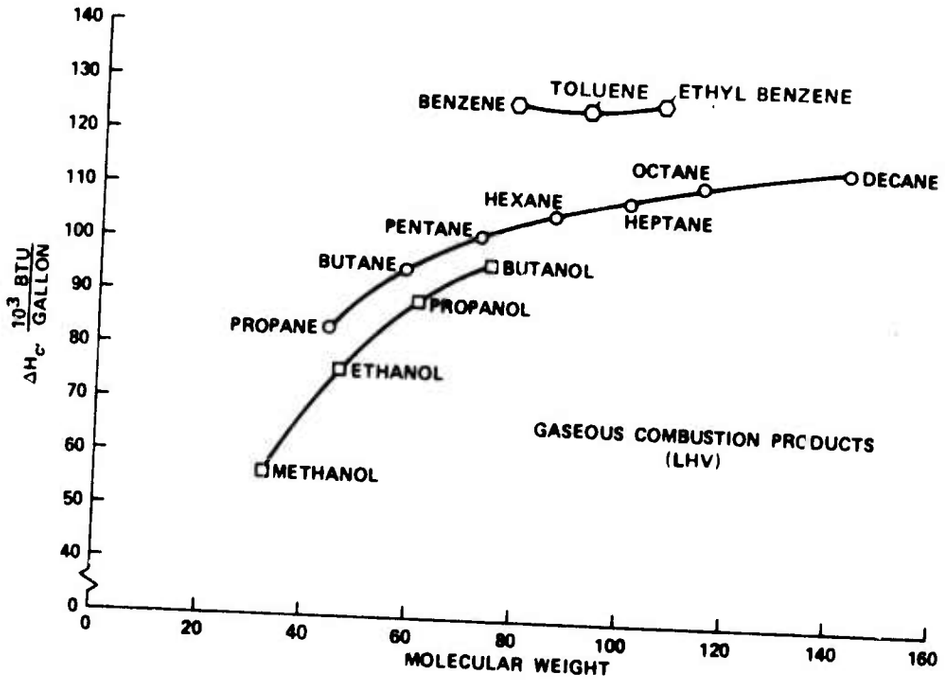

FIGURE 1. Volumetric Heat of Combustion Versus Molecular Weight.

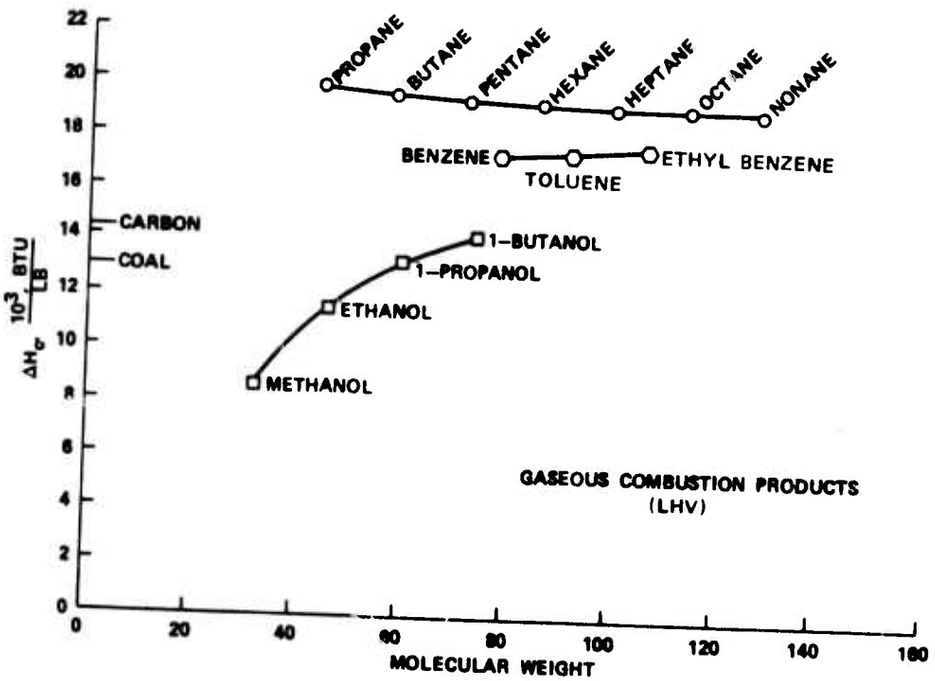

FIGURE 2. Gravimetric Heat of Combustion Versus Molecular Weight.

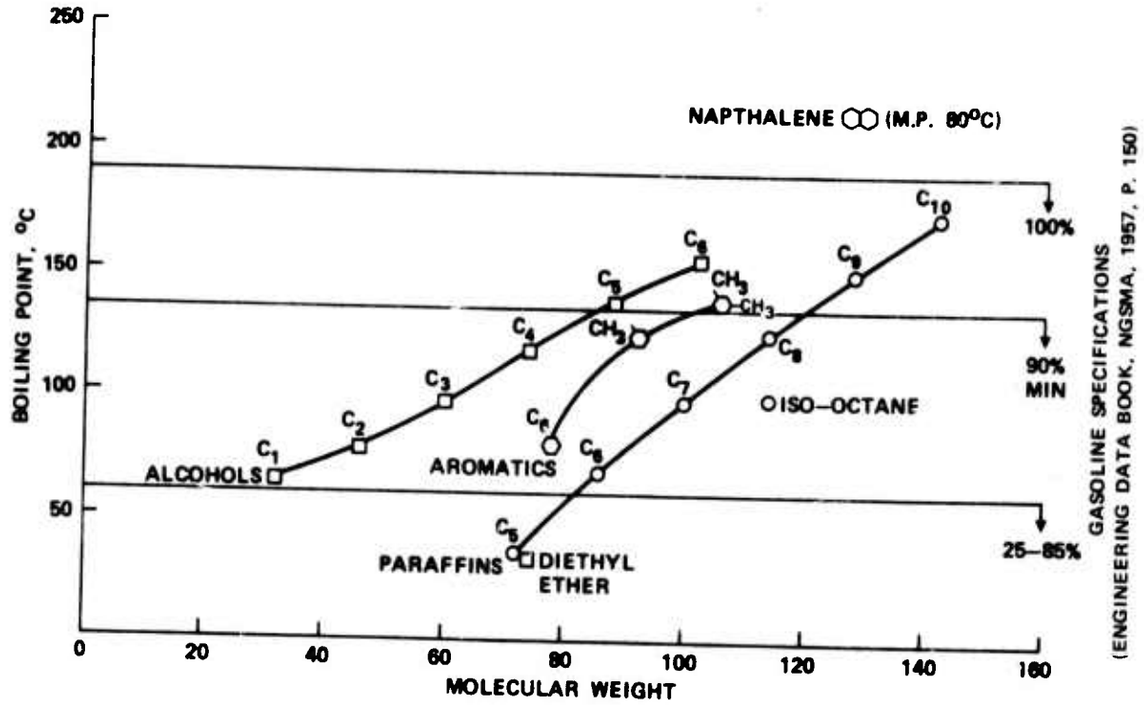

FIGURE 3. Boiling Point Versus Molecular Weight (Ref.: Lange's).

Four basic processes (physical, thermal, chemical, and biological) have been identified and are sufficiently distinct to permit differentiation as separate entities. These processes can be arranged in modular fashion, as shown in Figure 4, to show the sequence required to produce various fuel forms from solid waste.

A physical process can be simple (coarse shredding) or complex (separation of raw trash into various fractions of metals, glass, and organics with perhaps secondary and tertiary shredding of the organic material).

A thermal process involves addition of heat to bring about decomposition (pyrolysis) or combination (polymerization) reactions. If an intermediate chemical or catalyst is used to convert from one composition to another, the process is arbitrarily labeled chemical. An example of a chemical process is the catalytic synthesis of methanol from a mixture of hydrogen and carbon monoxide.

A biological process involves the action of enzymes or bacteria in converting from one chemical form to another. For example, the enzyme cellulase converts cellulose to glucose.

Of course, only a few of the many possible combinations of processes are shown in Figure 4, but many of the solid waste systems proposed and currently under development can be traced on this simplified chart.

TABLE 3. Properties of Candidate Fuels.

	Iso-octane	Methyl alcohol	Ethyl alcohol	Benzene
Chemical formula	C_8H_{18}	CH_3OH	C_2H_5OH	C_6H_6
Molecular weight	114	32	46	78
Carbon %, by weight	84.0	37.5	52.0	92.0
Hydrogen %, by weight	16.0	12.5	13.0	8.0
Oxygen %, by weight	Nil	50.0	35.0	Nil
Heating value				
Higher, Btu/lb	20,600	9,600	12,800	18,200
Lower, Btu/lb	19,100	8,650	11,500	17,470
Btu/gal (lower)	110,900	57,400	76,150	128,950
Latent heat of vaporization, Btu/lb	141	474	361	169
Specific gravity, 60°F	0.696	0.796	0.794	0.885
Stoichiometric mass A/F ratio	15.1	6.45	9.0	13.2
Boiling temperature, °F	211	149	172	176
Octane No., research	100	106	106	. . .
Octane No., motor	100	92	89	110
Density, lb/gal	5.8	6.6	6.6	7.4
Energy–Btu/lb air	1,265	1,340	1,280	1,320
Relative mileage (based on Btu/gal)	100%(ref.)	51%	68%	116%

TABLE 4. Theoretical Maximum Quantity of Fuels from One Ton of Dry Organic Solid Waste.

	Fuel, gal			
	Octane C_8H_{18}	Methanol CH_3OH	Ethanol C_2H_5OH	Benzene C_6H_6
Hydrogen limitation	147	164	157	240[b]
Carbon limitation	194	299	246	159
Energy limitation[a]	135	261	197	116

[a] 100% energy conversion.

[b] Additional carbon is required to use all of the available hydrogen.

Assumptions: Dry organic fraction has empirical formula: $C_6H_{9.1}O_{3.1}N_{0.13}S_{0.014}$

Lower heating value of organic fraction is 7,500 B/lb

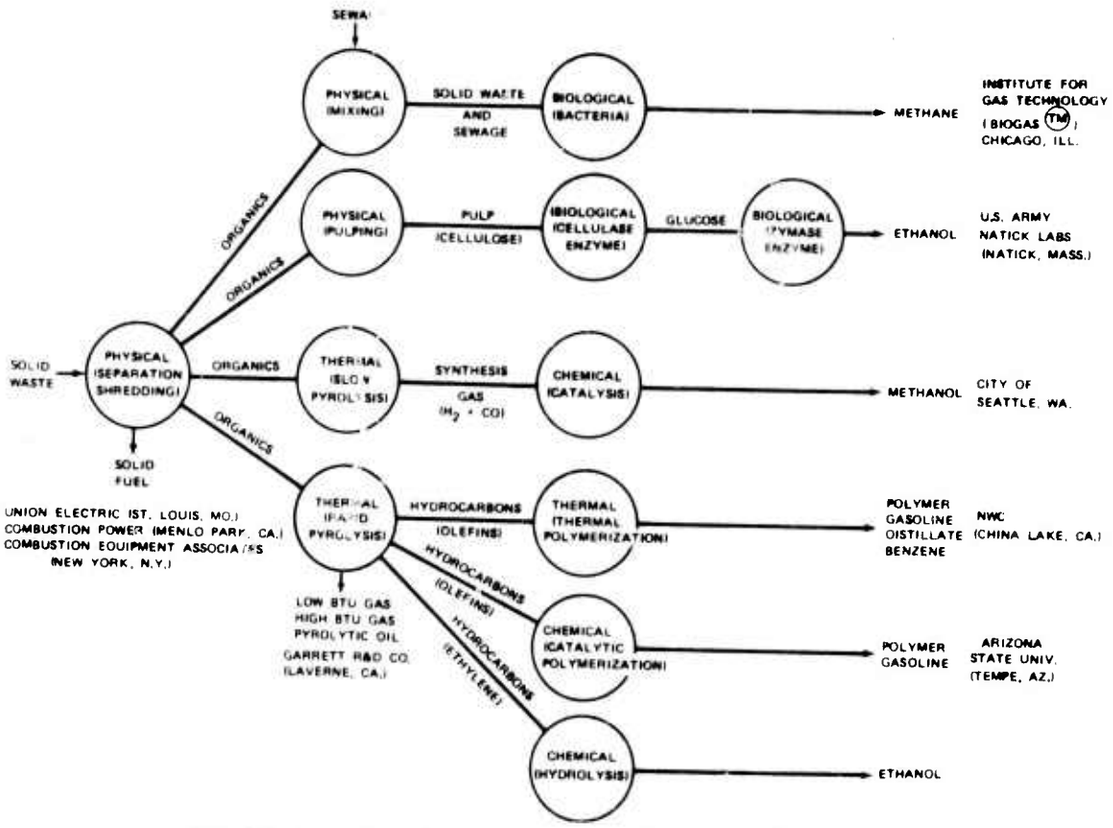

FIGURE 4. Process Routes to Alternate Fuels from Solid Waste.

PYROLYSIS

The liquid fuels being considered by this study all require a pyrolysis stage to convert the organic solid waste to either synthesis gas or hydrocarbon gases.

Pyrolysis of municipal and wood wastes has received considerable attention in the last few years[5-10] (see also the report referenced as footnote 1). Very slow pyrolysis processes, using a large particle size (about 1-inch) feed, yield products

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[5]Hamnond, V. L., et.al., "Energy from Solid Wastes by Pyrolysis-Incineration", Battelle Pacific Northwest Laboratories, Richland, Wa., November 1972.

[6]Burton, Robert S. and Richard C. Bailie, "Municipal Solid Waste Pyrolysis", West Virginia University. Paper No. 12D, AIChE 66th Annual Meeting, Philadelphia, Pa., November 1973.

[7]Brink, D. L., et.al., "A Flow Reactor Technique for the Study of Wood Pyrolysis", University of California Forest Products Laboratory, Berkley, Calif., October 1973.

[8]Mallan, G. M., et.al., "Gasification of Carbonaceous Solid". Garrett Research and Development Company. Patent No. 3,846,096, U.S. Patent Office, Patented 5 November 1974.

[9]Hammond, V. L. and K. L. Mudge, "Feasibility Study of Use of Molten Salt Technology for Pyrolysis of Solid Waste", Battelle Pacific Northwest Laboratories, Richland, Wa., April 1974.

[10]Naval Radiological Defense Laboratory. "Flash Pyrolysis of Solid-Fuel Materials by Thermal Radiation", by K. A. Lincoln. San Francisco, Calif., NRDL, 1965. Pyrodynamics, 2, 133-143.

containing small amounts of hydrocarbons and large amounts of char, water, hydrogen, and carbon monoxide. Rapid pyrolysis of small particle size feed (less than 1/8 inch) produces larger amounts of hydrocarbons at the expense of char, carbon monoxide, and hydrogen. The use of air or oxygen in the pyrolysis chamber, to partially oxidize the feed to supply the necessary heat of pyrolysis, appears to result in reduced hydrocarbon production. It is concluded that the products formed are more dependent on the method and condition of pyrolysis than on feed composition, as shown in Table 5.

The yields of the various components of dry organic solid waste are shown in Table 6. Based on this data, more realistic estimates were made on the upper limits of quantities of methanol, ethanol, gasoline, or benzene that can be obtained from solid waste. These estimates are shown in the last four columns of Table 6. Yields of methanol were based on the amounts of hydrogen and carbon monoxide available, assuming use of the water-gas reaction to obtain more hydrogen as necessary. Ethanol yields were based on the acid hydrolysis of ethylene.

Octane yields were based on the quantities of all hydrocarbon compounds, C_2 and higher. The constraint in this case was the hydrogen/carbon ratio and the amount of hydrogen available for these compounds. Yields for benzene were obtained in a similar manner. It is apparent that the constraint of using pyrolysis products considerably reduces the amount of liquid fuels that can be realistically obtained from solid waste.

SYNTHESIS

Based on the above estimated yields, the two most promising solid waste fuel derivatives are methanol and polymer gasoline. The processes for conversion to these fuels were studied in more detail and some of the findings are presented below.

Methanol

Mass and energy balances of two processes for making methanol were carried out. The flow sheets for the two processes are shown in Figures 5 and 6. Both processes use Imperial Chemical Industries low-pressure (750 psi) process to catalytically form methanol. However, they differ radically in the pyrolysis and synthesis gas preparation steps. The low-pressure process was chosen for study because it has a lower compression energy requirement than high-pressure processes.

The amount of methanol in the synthesizer gas output is on the order of 2.5%. This requires recycling a large amount of synthesis gas per pound of methanol formed (about 17:1). Any inert gases, such as methane or nitrogen, slowly accumulate and decrease the partial pressure of the reacting gases, thus decreasing methanol formation.

TABLE 5. Pyrolysis Product Composition Comparison.

Agency	Ref.	Type of reactor	Dried gas composition, mole %								Dry mol. wt.	Percent gassified	Temp., °F
			H_2	CO	CO_2	CH_4	C_2H_2	C_2H_4	C_2H_6	C_{3+}			
Battelle	5	Vertical shaft w/air	38.1	37.1	20.8	3.2	. . .	0.48	0.26	. . .	21.0	100	1,330
BuMines	1	Retort (batch)	51.9	18.2	11.4	12.7	. . .	4.68	0.14	1.0	15.1	76	1,650
W. Va. Univ.	6	Fluidized sand	44.0	22.6	18.0	5.6	. . .	1.6	0.70	0.94	18.6	92	1,460
U.C. Berkley	7	Fluidized feed w/N_2	17.0	54.6	7.6	16.2	. . .	4.5	0	. . .	22.8	90	1,550
Garrett	8	Fluidized char	16.7	17.9	23.1	15.4	. . .	20.9	1.3	4.9	27.1	86	1,400
Battelle	9	Molten salt (batch)	32.4	31.8	9.9	18.3	. . .	6.5	1.0	0.05	19.0	54	1,650
USNRDL	10	Xenon flash radiation	0	52	4	0	31[a]	13[a]	0	0	28.0	99	1,100

[a]From mass and elemental balances for volatile organic fraction.

TABLE 6. Typical Yields of Pyrolysis Products and Their Maximum Potential for Fuels Per Ton of Dry Organic Material.

Agency	H_2, lb	CO, lb	CO_2, lb	CH_4, lb	C_2H_2, lb	C_2H_4, lb	C_2H_6, lb	C_{3+}, lb	MeOH, gal	EtOH, gal	Octane gasoline, gal	Benzene, gal
Battelle	73	989	872	49	0	13	0	0	115	3	2	2
Bu Mines	104	513	505	205	0	132	4	58	88	33	32	24
W. Va. Univ.	87	626	783	89	131	44	21	54	106	11	32	32
U.C. Berkley	27	1,207	264	205	0	99	0	0	91	25	15	12
Garrett	21	318	645	156	0	371	25	180	35	93	97	71
Battelle	37	506	248	166	0	103	17	2	59	26	20	15
USNRDL	0	740	80	0	417[a]	183[a]	0	0	0	46	103	80

[a]From mass and elemental balances for volatile organic fraction.

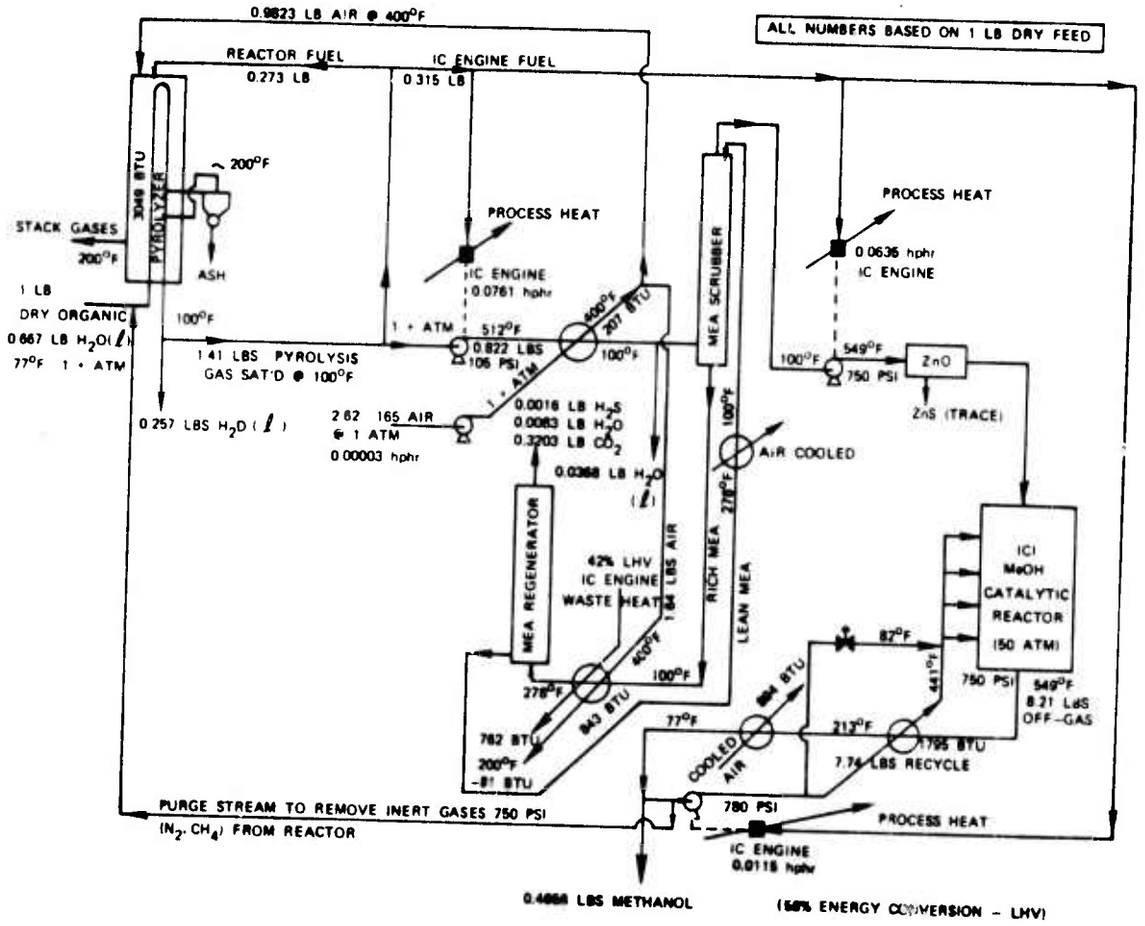

FIGURE 5. Methanol Flow Sheet No. 1.

Consequently, it is very important to minimize the amount of inert gases in the synthesis gas stream. The inert gases are maintained at an acceptable level by purging. Side reactions are minimized by the presence of excess hydrogen.[11]

The overall chemical reaction in the conversion of cellulosic materials to methanol can be represented as

$$C_6H_{10}O_5 + 3H_2O_{(\ell)} \rightarrow 4CH_3OH_{(\ell)} + 2\ CO_2 \quad \text{(endothermic)} \qquad (1)$$

or

$$C_6H_{10}O_5 + 9/4\ O_2 \rightarrow 2\ 1/2\ CH_3OH_{(\ell)} + 3\ 1/2\ CO_2 \quad \text{(exothermic)} \qquad (2)$$

[11] Danner, G. A. (ed.), *Methanol Technology and Economics*, Chemical Engineering Progress Symposium Series, Vol. 66, No. 98, p. 29, 1970.

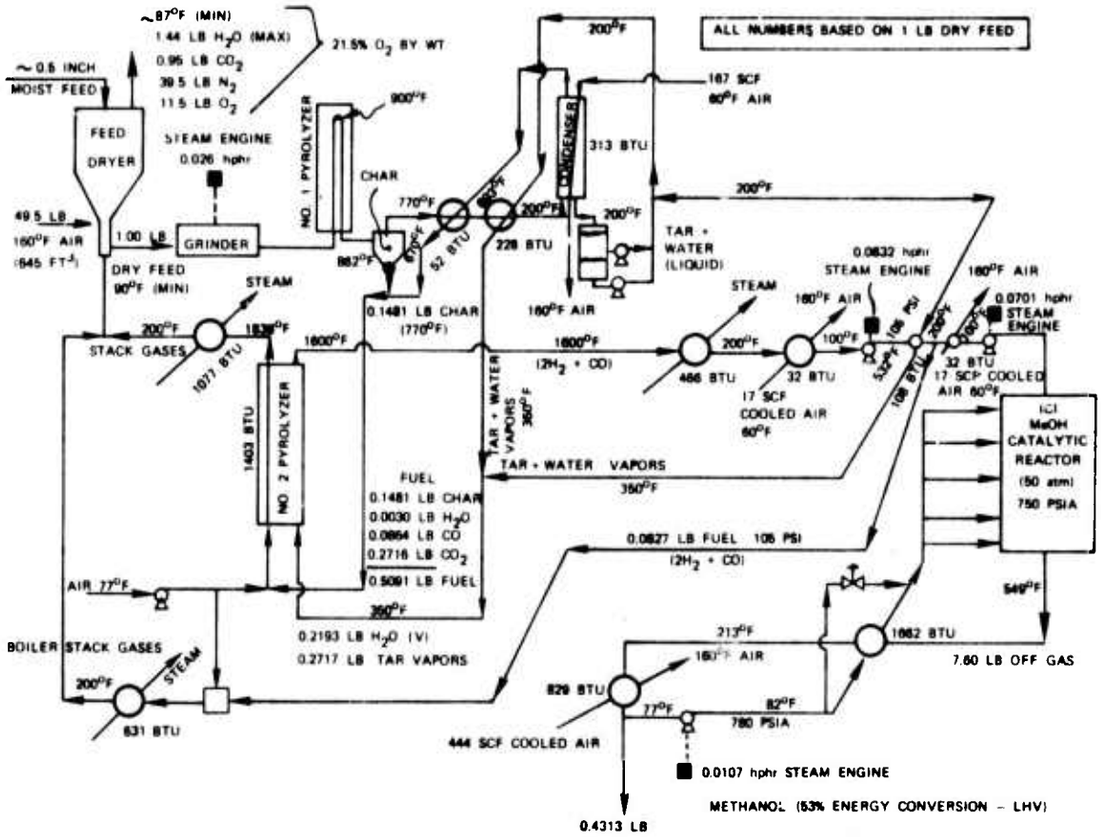

FIGURE 6. Methanol Flow Sheet No. 2.

There is a shortage of hydrogen in the process since cellulose has a hydrogen-to-carbon molar ratio of 1.67:1, while that of methanol is 4:1. Assuming the process must supply the hydrogen from water, carbon is consumed by the water-gas reaction

$$C + 2\ H_2O \rightarrow CO_2 + 2H_2 \qquad (3)$$

These considerations lead to the conclusion that there will be excess carbon dioxide which, if not removed from the synthesis gas stream, will accumulate in the methanol synthesizer where it will promote side reactions. Accumulation of carbon dioxide will also decrease the methanol yield, since the equilibrium methanol composition increases with the cube of the hydrogen partial pressure, but only linearly with the carbon dioxide partial pressure. Since the compression of the synthesis gas consumes a very large amount of energy, it is imperative that the excess carbon dioxide be removed at the lowest economically feasible pressure. The major difference between the two methanol flow sheets is the method used to remove this excess carbon dioxide.

In flowsheet 1 (Figure 5), the cellulosic materials are pyrolyzed in the presence of water to produce an excess of two moles of carbon dioxide for every four moles of methanol (Eq. 1). The pyrolysis gases are cooled to 100°F to condense out some of the water, then split into three streams: (1) pyrolysis reactor fuel (19%), (2) internal combustion (IC) engine fuel (22%) for process shaft work, and (3) synthesis gas (59%). The synthesis gas is then compressed to 105 psig and passed through a monoethanolamine (MEA) scrubber to remove excess carbon dioxide and a large portion of the hydrogen sulfide. Following this, the synthesis gas stream is compressed to 750 psi, passed through a final hydrogen sulfide removal process (ZnO), and passed into the methanol synthesizer. The synthesizer is operated at 750 psig, 549°F, and a recycle ratio of 17 pounds per pound of methanol. The recycle compressor consumes 8% of the total compression power required. This recycle power is lower (percentagewise) than reported (see footnote 11) but reflects the lower initial pressure of the synthesis gas in Figure 5 and the subsequent higher compression power requirements. The shaft power is obtained from IC engines operating at an assumed 25% conversion of the lower heating value (LHV) to shaft work. Low temperature energy is recovered from the IC engines (42% LHV)[12] and the interstage compressor cooler, and then used to heat the MEA stripping column. This process will produce 0.47 pound of methanol per pound of dry cellulosic feed material and is independent of outside energy sources. This is an energy conversion of 58% of the cellulose LHV. The 42% waste energy is in the form of clean, hot (180-200°F) air which could be used for space heating and cooling of office complexes or apartments (assuming proper location of the methanol plant).

The second methanol flow sheet (Figure 6) uses a novel two-stage pyrolysis to effect a carbon dioxide separation. The overall chemical reaction can be visualized in Eq. 2. The cellulosic feed is dried and ground to a very fine particle size. First-stage pyrolysis is then accomplished at a relatively low temperature (900°F) with a very short residence time. This has been shown to produce a large amount of char, carbon dioxide, and pyrolytic oil, but only small amounts of water, hydrogen, and carbon monoxide[13] (see also the report referenced as footnote 7).

The char is removed from the gas stream via a cyclone separator. The pyrolytically formed oil and water are condensed from the gas stream and stored. The non-condensable gas stream is recombined with the char and used to convey the char to the second-stage pyrolysis unit where it is burned for process energy. The condensed pyrolytic oil and water are then individually metered into the second-stage pyrolysis unit where they are subjected to 1,600°F for several seconds at about one atmosphere pressure.

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[12]Perry, Robert H. (ed.), *Chemical Engineer's Handbook*, 4th Edition, New York, McGraw-Hill, p. 24-68, 1963.

[13]Finney, C. S., "New Techniques in the Pyrolysis of Solid Wastes", Garrett Research and Development Company 72-035 AIChE 73rd National Meeting. August 1972.

The heat transfer medium in the second-stage pyrolysis unit is molten sodium carbonate, which catalyzes the required water-gas reaction and chemically reacts with and removes any hydrogen sulfide present in the synthesis gas stream. Since the pyrolytic oil has a very low ash content, the immense carbonate recovery problem encountered with pyrolyzing high-ash-content raw cellulose is eliminated (see footnote 9). Although it would be tempting to operate this second-stage pyrolysis unit at elevated pressures to avoid compression of synthesis gases, higher pressures would favor the formation of some methane rather than the desired synthesis gas. Any methane gases formed by the second-stage pyrolysis of the pyrolytic oil and water, are compressed to 750 psi and passed into the methanol synthesizer. Methanol is formed as previously discussed (see Figure 5). The hot stack gases and the second-stage pyrolytic gases are passed through heat exchangers to make steam. This 100 psig steam with 100°F superheat (435°F steam) is used to run small steam engines operating at a 20% conversion of thermal energy to shaft energy. Steam rather than turbine engines were chosen because of the low efficiencies of turbine engines using steam at pressures below 300 psig (see pp. 24-74 of the report referenced as footnote 12). Again, waste heat could be utilized as hot air for space heating and/or cooling.

The 200°F stack gases are used to dry the feed. Feed containing as much as 58% by weight moisture can be dried in this manner. A small portion of the synthesis gas (0.06 pounds per pound of dry feed) is burned to provide additional process energy. For every pound of dry cellulosic material, 0.43 pound of methanol is produced. The energy conversion of this process is 53% based on the LHV.

Polymer Gasoline

Nearly all research in pyrolysis has shown that unsaturated hydrocarbons (olefins) are present to some extent in the pyrolysis gases. The amount of all hydrocarbons, other than methane (C_2+), is calculated to vary from 13 (see footnote 5) to 600 (see footnote 10) pounds per ton of dry organic material. Very rapid pyrolysis at atmospheric pressure and high temperatures favors the formation of low molecular weight hydrocarbons and carbon dioxide, while at the same time decreasing the formation of hydrogen, carbon and water. Detailed chemical analysis of the C_2+ hydrocarbons formed by pyrolysis shows them to be 84% by weight olefins (see footnote 1). This high degree of unsaturation is predicted from the relatively higher thermal stability of unsaturated compared to saturated hydrocarbons.[14]

This large olefin content in the hydrocarbon fraction of the pyrolysis gases could be utilized to make what the petroleum industry refers to as polymer gasoline. The basic research of polymerizing low molecular weight hydrocarbons to gasolines was performed in the 1930s. This research led to the successful commercial utilization of the low molecular weight unsaturated hydrocarbons formed as a by-product in the pyrolysis (cracking) of crude oil to gasolines.

[14]Nelson, W. L., *Petroleum Refinery Engineering*, 4th Edition, New York, McGraw-Hill, p. 628, 1958.

The two major commercial processes for polymerizing the low molecular weight olefins are non-catalytic (thermal) and catalytic (phosphoric acid). The phosphoric acid process has the advantages of lower pressure and lower temperature, producing a gasoline having a motor octane of 84 and a blending octane value (BOV) of 115, but it converts hydrogen sulfide to mercaptans and polymerizes ethylene with relative difficulty.[15] The thermal process is more tolerant of hydrogen sulfide, preferentially polymerizes ethylene[16] and normally produces gasoline having a slightly lower motor octane of 78 and a BOV of 87 (see p. 725 of report referenced as footnote 14), but it can be operated at higher temperatures to form a more aromatic gasoline with an octane rating of 100.[17] The ability of the process to polymerize ethylene is quite important, since it is estimated that ethylene comprises 65-83% of the olefins formed by pyrolysis of trash.

In pyrolysis gases formed at 1,650°F, C_2+ saturated hydrocarbons were ethane at 2.2% and butane at 13.6% by weight of the hydrocarbon fraction (see p. 10 of report referenced as footnote 1). These saturated gases will tend to accumulate in the polymerizer, although varying amounts of the butane are acceptable in the gasoline product, depending on the season. The saturated hydrocarbons may be converted to olefins by pyrolysis at atmospheric pressure and about 1,550°F, and a very short contact time (on the order of less than 4 seconds). Pyrolysis at elevated pressures reduces the olefin yield.

The pyrolysis of ethane can result in a 90% by weight yield of ethylene based on the ethane consumed. The side reactions during the cracking of ethane produce methane (8%) and, to a lesser degree, acetylene (2%). The yield of olefins from the pyrolysis of butane at one atmosphere and 1,300°F is also about 90% by weight, although the olefin yield is considerably less at higher temperatures.[18]

The polymerization of olefins can proceed at atmospheric pressure and at temperatures on the order of 1,500°F.[19] At this temperature, the polymers formed are unstable and rapidly decompose to form naphthenic tars and char by the elimination of hydrogen. Polymerization under these conditions results in a product that is about half aromatic gasoline with a boiling point below 392°F (75% benzene) and a

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[15] Ipatieff, V. N., et.al., "Polymerization, A New Source of Gasoline", Industrial and Engineering Chemistry, Vol. 27, No. 9, p. 1077-1081 (1935).

[16] Sullivan, F. W., Jr., et.al., "Pyrolysis and Polymerization of Olefins", Industrial and Engineering Chemistry, Vol. 27, No. 9, p. 1072-1076 (1935).

[17] Wagner, C. R., "Production of Gasoline by Polymerization of Olefins", Industrial and Engineering Chemistry, Vol. 27, No. 8, p. 936 (1935).

[18] Frolich, K., et.al., "Cracking and Polymerization of Low Molecular Weight Hydrocarbons", Industrial and Engineering Chemistry, Vol. 27, No. 9, p. 1055-1062 (1935).

[19] Dunstan, A. E., et.al., "Thermal Treatment of Gaseous Hydrocarbons, I. Laboratory Scale Operation", Industrial and Engineering Chemistry, Vol. 26, No. 3, p. 307-314 (1934).

higher-boiling-point tar fraction containing large amounts of naphthenic hydrocarbons. It is recommended that hydrogen sulfide levels be kept below 1% to minimize the formation of rubbery by-products.[20]

Lower temperatures may be used to reduce the formation of tars, but higher pressures are needed to maintain the reaction rates. At 850°F and 500 psi, both ethylene and propylene polymerize to form liquids that are 80% gasoline with a CFR Research octane rating of about 80. The rate of ethylene polymerization is roughly twice that of propylene at these conditions. Increasing the pressure greatly increases the rate of polymerization, at the expense of gasoline fraction and octane rating. For example, with ethylene, by increasing the pressure from 500 to 3,000 psi, the rate of liquid formation at 850°F is increased by a factor of about 10. However, the gasoline in the liquid product drops from about 75% to about 65%, and the octane rating drops from 78 to 63. The major advantage of the high pressure is a large reduction in the size of the polymerizer unit (see p. 1076 of report referenced as footnote 16). With a mixture of two parts ethylene to one part propylene, a single-pass conversion of 78% of the olefins to liquids was achieved at 600 psi and 750°F. These liquids all boiled below 437°F (see p. 1059 of report referenced as footnote 18). The 10-90% boiling range is about 115-350°F for gasolines and about 385-475°F for JP-5 (see p. 34-49 of report referenced as footnote 14).

The trash pyrolysis gases containing the highest reported hydrocarbon content were chosen to evaluate the feasibility of making polymer gasoline. Although this flow sheet is not complete, some preliminary findings are worth discussing. Based on the C_2+ hydrocarbons being separated from the pyrolysis gases at high pressure and low temperature, it was calculated that more than sufficient energy was obtainable from combustion of the remaining gases to provide the heat of pyrolysis at a stack temperature of 1,800°F. In addition, there was more than sufficient fuel to use IC engines to power the shredder and to compress all the pyrolysis gases to 750 psi. The compression power requirements are considerably less for making polymer gasoline than methanol, since the energy required to compress the gases is roughly proportional to the volume or moles of gases compressed per weight of product. Compressing 1 mole of ethylene produces 28 pounds of polymer gasoline; compressing 1 mole of synthesis gas yields only 11 pounds of methanol.

The methanol flow sheets involved a tradeoff between the amounts of synthesis gas used to meet process energy requirements and to produce methanol. However the polymer gasoline process apparently has a relatively lower power requirement and production will be constrained by the quantity of hydrocarbons in the pyrolysis gases. Based on this, at a 90% conversion of paraffins to olefins and olefins to polymers (see

[20]Cadman, W. A., "Thermal Treatment of Gaseous Hydrocarbons, II. Semi-Industrial Production of Aromatic Hydrocarbons from Natural Gas in Persia", Industrial and Engineering Chemistry, Vol. 26, No. 3, p. 315-320 (1934).

p. 1059 of report referenced as footnote 18), the prediction for polymer gasoline production is 0.18 pound of gasoline and 0.06 pound of fuel oils per pound of dry organic material (or 62 and 19 gallons per ton, respectively). This represents an energy conversion of 66% based on the LHV.

FLOW SHEET COMPARISON

The major difference between the two methanol flow sheets is the method of rejecting excess carbon dioxide from the synthesis gas. Both flow sheets resulted in nearly the same energy conversion, 58% versus 52%. This small difference is probably not significant in view of some of the uncertainties in the assumptions, e.g., the pyrolysis assumptions need further experimental verification. Major factors in the methanol process inefficiency are (1) the highly endothermic heat of pyrolysis that is not recoverable as process heat in the exothermic methanol synthesis step, and (2) the very large power requirement for compression of the synthesis gases. At this time it appears the single-stage pyrolysis with scrubbing to remove excess carbon dioxide is more straightforward than the two-stage pyrolysis process.

In contrast to the methanol flow sheet, the preliminary polymer gasoline flow sheet shows that (1) the desired pyrolysis to a large percentage of hydrocarbons requires less energy input than the pyrolysis to the minimal percentages desired for the methanol synthesis, and (2) less compression power is needed for polymer gasoline. For the assumed pyrolysis gases, the polymer gasoline process is limited by the C_2+ hydrocarbons content and not by process energy requirements (assuming that pyrolysis and compression are the major energy sinks as they were for the methanol process). The polymer gasoline process, although not finalized, appears to have an energy conversion of 66%.

The preliminary polymer gasoline flow sheet is more promising than methanol in that a 14-25% better energy conversion is predicted. Since polymer gasoline is a mixture of hydrocarbons, it would introduce no new corrosion problems and would be unquestionably interchangable with other gasolines. However, it should be pointed out that in engines with low efficiencies (about 14%), up to 30% methanol may be added to gasoline with no decrease in miles per gallon (MPG) of fuel,[21] although other research has shown that a decrease in mileage can occur with certain engines as the alcohol content is increased[22] (see also footnotes 3 and 4). In a blending application of the synthetic product, a gallon of methanol may give as many miles as a gallon of gasoline. On a volume basis, 81 gallons of polymer gasoline per ton are to be

[21] Naval Weapons Center. *Compilation and Analysis of Test Data from an Internal Combustion Engine Utilizing Gasoline-Methanol Fuel Mixtures*, by D. R. Cruise. China Lake, Calif., NWC, 13 June 1974. (NWC Reg. Memo 451-202-74.)

[22] Wigg, E. E., "Methanol as a Gasoline Extended: A Critique", Science, 186, 785-790 (1974).

compared to about 136 gallons of methanol per ton, or 68% more gallons of methanol than polymer gasoline. It becomes very clear that if capital and operating costs for methanol and polymer gasoline are comparable, then selection of one process over the other would be based on the intended usage of the fuel. For methanol the intended use would be to extend gasoline supplies by blending. Polymer gasoline and the by-product fuel oils would provide overall fuel system compatibility with no question of separation into gasoline and aqueous alcohol phases and would unquestionably maximize vehicle range per pound of fuel.

The synthesis of both methanol and polymer gasoline is an accomplished and commercialized fact, although from other synthesis gas sources. However, the flow sheet development has served to emphasize the need to verify certain reported pyrolysis results.

PRELIMINARY ECONOMIC ANALYSIS

Preliminary economic analyses were performed for a solid waste facility having (1) a front-end system for the separation of iron, aluminum, heavy metals, and glass, (2) a pyrolysis system to gasify the organic fraction of the trash, and (3) a synthesis system to produce methanol suitable for use as a fuel. The chemical and petroleum industries have established that, for preliminary economic analyses, prediction of new plant cost is best based on previous cost of similar plants adjusted for differences in plant sizes and inflation. A general rule of thumb is that the ratio of various plant costs is equal to the ratio of plant sizes raised to an exponent of 0.5-0.9.[23,24] An exponent of 0.65 was chosen for plant cost extrapolation. The use of too small an exponent results in slightly higher estimated costs when extrapolating down to the smaller plant sizes of interest. As with all extrapolations, there is less possible error with less extrapolation. The effect of inflation on the cost of building chemical plants is adjusted through special cost indices developed for just such a purpose. The Chemical Engineering Plant Index was used. (The Nelson Refinery Construction Index or the M and S Equipment Cost Index could have been selected instead with very small changes in the results of the analysis.)

The front-end system chosen for extrapolation was developed by a current state-of-the-art review and does not actually exist at this time. The hypothetical system would process 500 tons of raw trash per day.[25] The value of reclaimed materials is

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[23]Peters, M. S., et.al., *Plant Design and Economics for Chemical Engineers*. New York, McGraw-Hill, p. 107. (1968)

[24]Miller, C. A., "Current Concepts in Capital Cost Forecasting", Chemical Engineering Progress, Vol. 69, No. 5, p. 79. (1973)

[25]Abert, James G., et.al., "The Economics of Resource Recovery from Municipal Solid Waste", Science, 183, p. 1052-1058. (1974)

skyrocketing due to shortages of raw materials and fuel, but for this analysis the following values were assumed: the raw trash is 5.5% iron valued at $40 per ton; 0.5% aluminum at $300 per ton; 6.6% glass at $7 per ton; 0.2% other nonferrous metals at $120 per ton; and 27% moisture. No credit was taken for dumping fees, since this is extremely variable from site to site. (The reader can easily take a specific dump fee into account, as will be discussed later.)

The pyrolysis system chosen for extrapolation is based on a Stanford Research Institute analysis[26] of West Virginia University's pyrolysis system (see footnote 6). Their analysis was for a plant size capable of processing 1,358 tons of raw trash per day (1,000 tons dry raw trash per day). The methanol synthesis section is based on the construction and operating cost of a plant capable of producing 500 tons of methanol per day, with an adjustment made for not needing a methane reformer nor a distillation column due to the relatively pure methanol product. This methanol synthesis section uses the ICI methanol technology and cost data.[27]

The results of the economic analyses are shown graphically in Figures 7-13, while the detailed tabulated bases are shown in Table 7. The plant site is assumed to operate 350 days per year. The effect of population size on the operating costs of a complete trash-to-methanol plant is shown in Figure 7 for various methanol values. As would be expected, the larger the population base (plant size), the lower the operating costs (or the greater the profit). The operating costs become much more sensitive for population sizes below about 60,000 since the minimum personnel force that can operate the plant has been reached. The operating costs take credit for the assumed values of recovered iron, aluminum, heavy metals, and glass and for the methanol produced, but do not take credit for a dumping fee. If the plant is paid a dumping fee of two dollars per ton to dispose of the trash for 25,000 people, the break-even point (or zero cost) would be adjusted upward and result in the methanol costing $0.38/gallon, rather than $0.40/gallon without the dumping fee.

In Figure 8, operating costs are plotted using the energy value of the methanol as a parameter. For comparison, the delivered cost of gasoline to NWC in August 1974 was $0.42/gallon or about $3.65/MBtu.

Due to the inherently higher yield per pass and smaller compressor loads of the thermal polymerization process used in the polymer gasoline flow sheet, the capital costs are expected to be about 20% lower than for the methanol system. Operating costs for the polymer gasoline process (Figures 9 and 10) were based on this assumption.

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[26] Alpert, S. B., et.al., "Pyrolysis of Solid Waste: A Technical and Economic Assessment", Stanford Research Institute, Menlo Park, Calif., September 1972.

[27] Kenard, R. J., et.al., "Present Methanol Manufacturing Costs and Economics Using the ICI Process", *Chemical Engineering Progress Symposium* Series No. 98, Vol. 66, AIChE, p. 47-53. (1970)

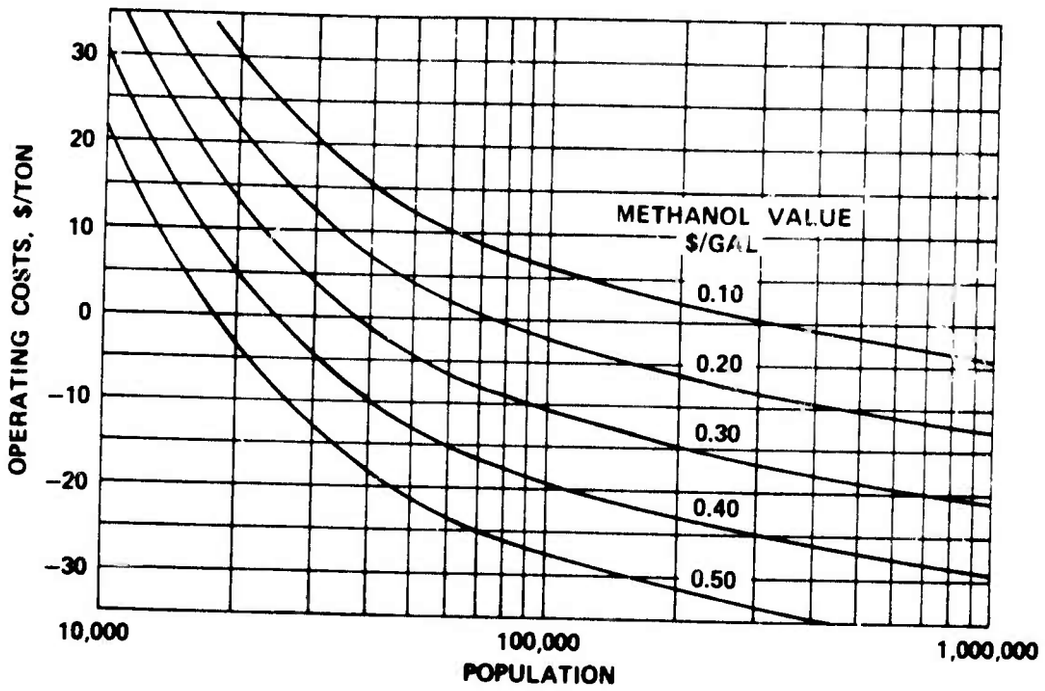

FIGURE 7. Net Operating Costs of Solid Waste Facility with Material Recovery and Conversion of Organic Fraction to Methanol.

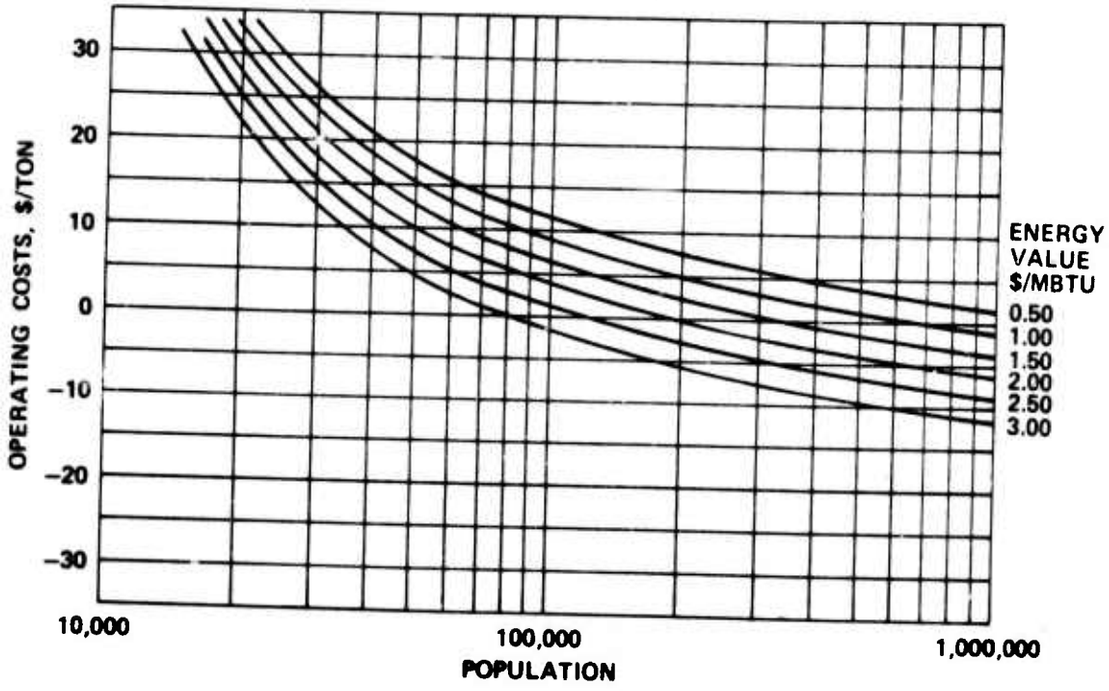

FIGURE 8. Net Operating Costs of Solid Waste Facility with Material Recovery and Conversion of Organic Fraction to Methanol.

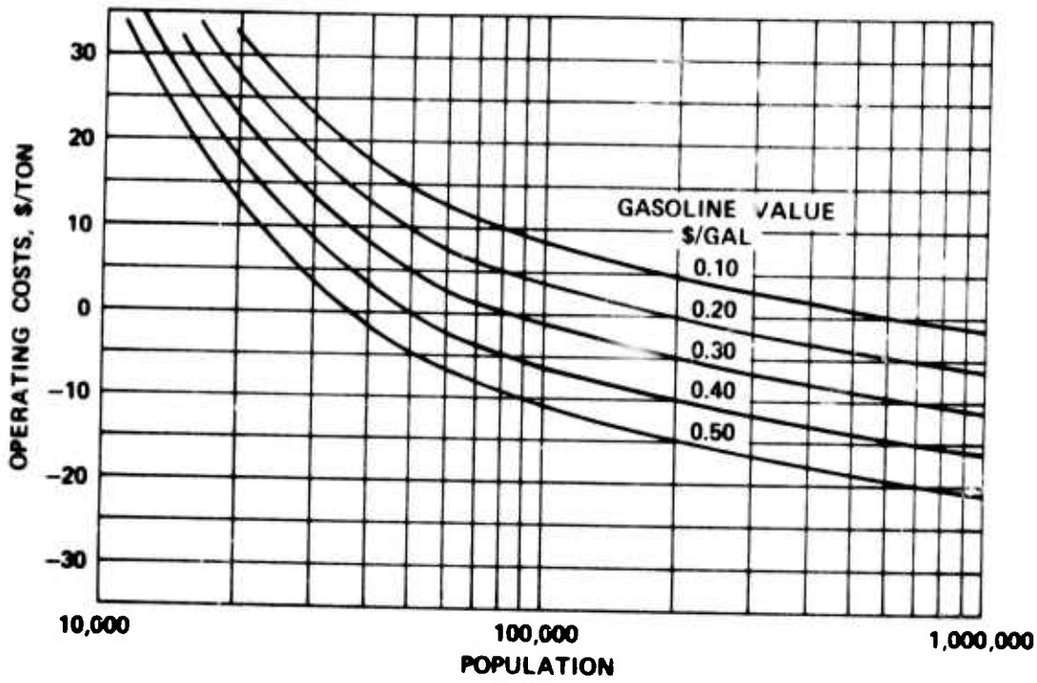

FIGURE 9. Net Operating Costs of Solid Waste Facility with Material Recovery and Conversion of Organic Fraction to Polymer Gasoline.

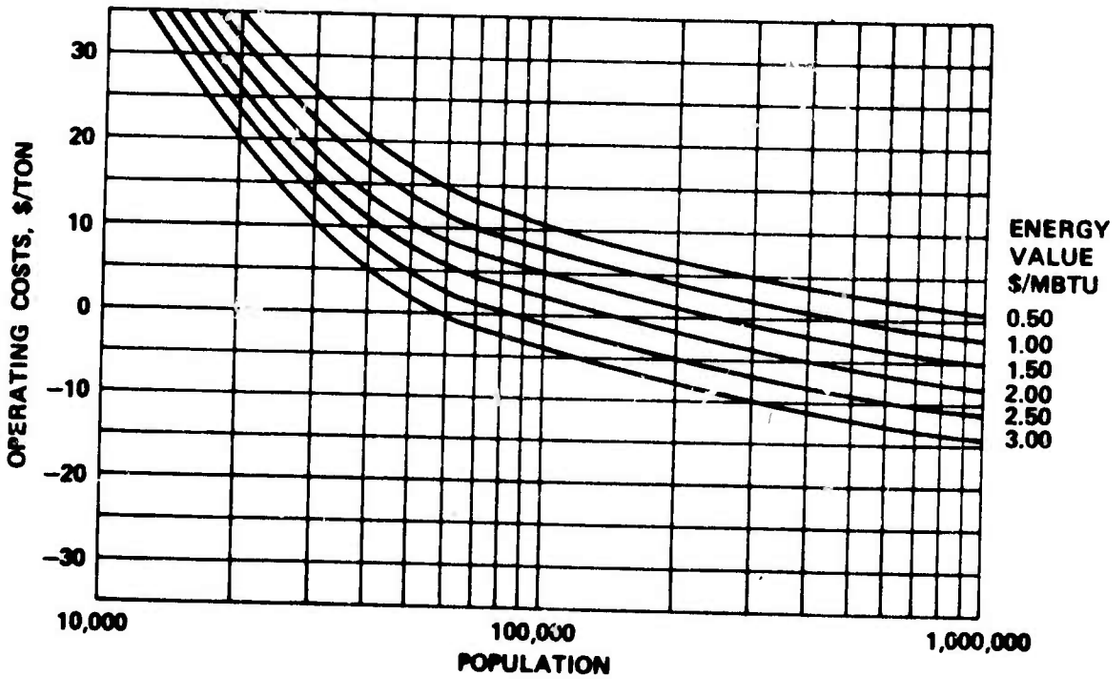

FIGURE 10. Net Operating Costs of Solid Waste Facility with Material Recovery and Conversion of Organic Fraction to Polymer Gasoline.

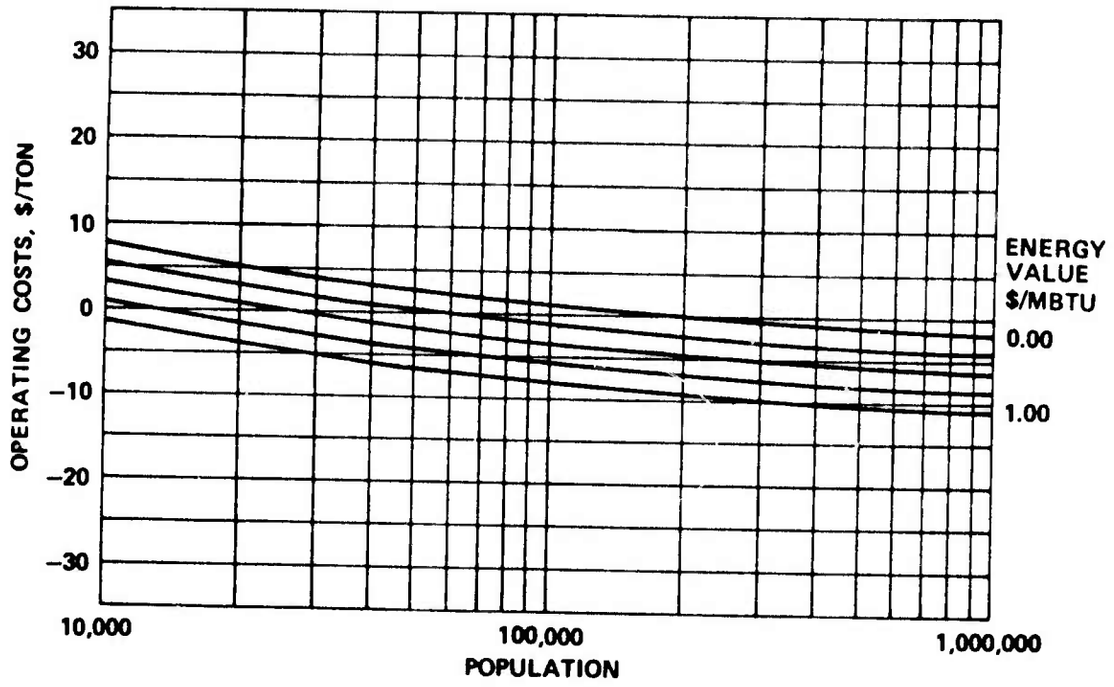

FIGURE 11. Net Operating Costs of Solid Waste Facility with Material Recovery and Use of Organic Fraction as Substitute Coal.

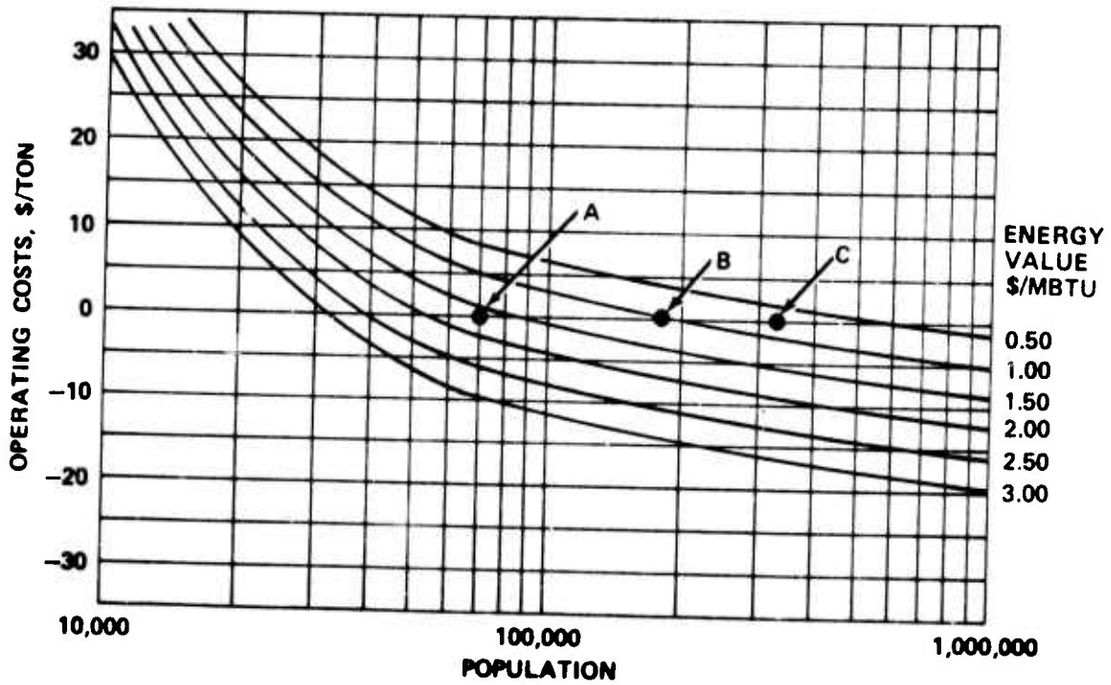

FIGURE 12. Net Operating Costs of Solid Waste Facility with Material Recovery and Conversion of Organic Fraction to Oil or Gas.

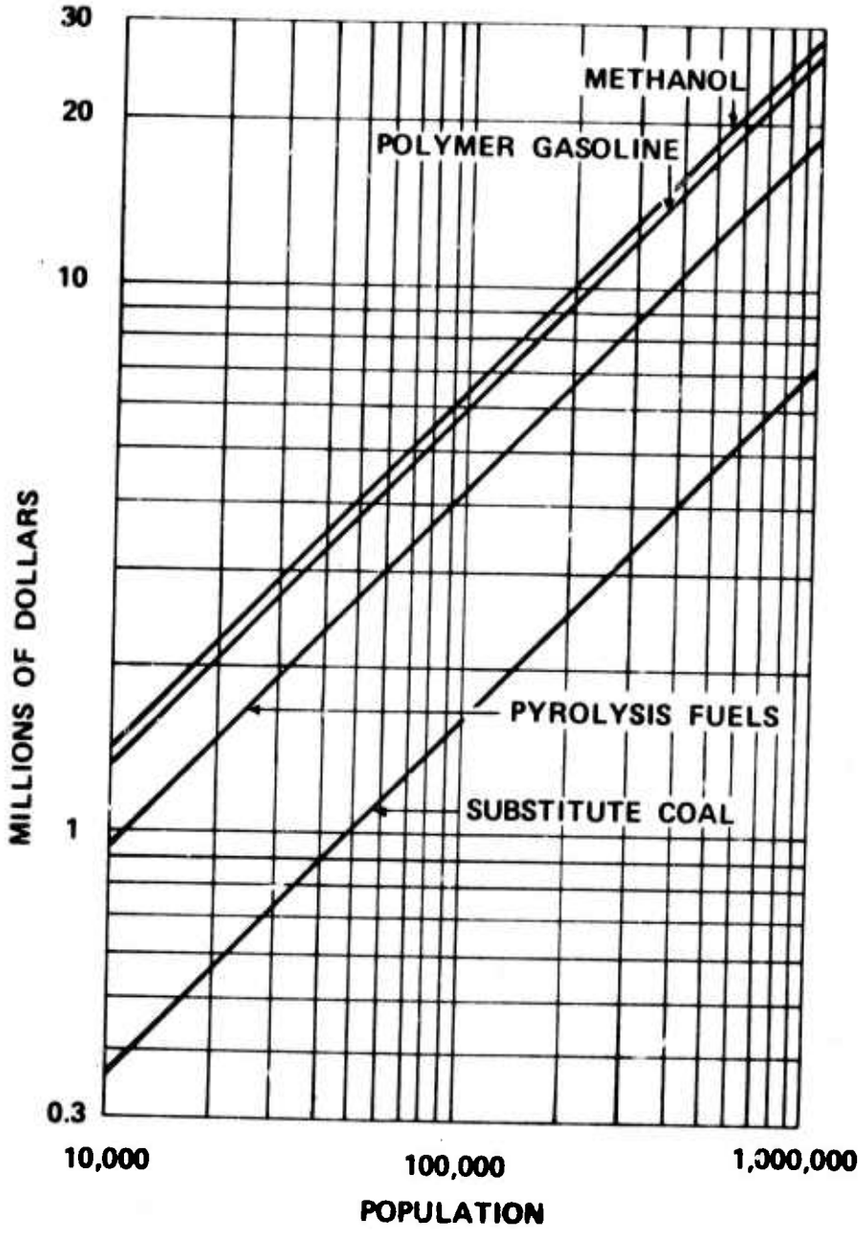

FIGURE 13. Effect of Population on Total Capital Costs of Solid Waste Conversion Facilities.

TABLE 7. Explanation of Plant-Size Scaling Bases.

Item	Front-end system[a]	Pyrolysis unit[b]	Imperial Chemical Industries' methanol synthesis[c]
Total capital investment (TCI), millions of dollars	2.42	7.49	7.34
Capacity (raw trash), tons/day	500	1,358	1,812 (500 MeOH)
Cost basis year	1974	1972	1970
Operating days per year	312	312	350
Annual operating costs, % of TCI			
Labor, L	0.12	0.096	0.02
Utilities, U	0.013	0.0022	0.0027
Materials, M	0.031	0.029	0.032
General overhead = 1/2(L+M)	0.075	0.0625	0.026
Fixed costs			
Amortization of TCI over 25 years at 6.5% interest	0.082	0.082	0.082
Total operating costs/TCI	0.321	0.272	0.163

[a]See footnote 25.
[b]See footnote 26.
[c]See footnote 27.

As expected, methanol has a lower net operating cost than polymer gasoline on a volume basis, while polymer gasoline appears more attractive on an energy basis. This type of comparison highlights the need to determine the overall system feasibility of using methanol to extending gasoline supplies.

The net operating costs for a substitute coal fuel are shown in Figure 11. This system consists of only a front-end system to separate the organic fraction from the metals and glass and shred to a particle size that can be conveniently fed into a boiler. Combustion Equipment Associates, Inc. of New York markets a confetti-type solid waste called ECO-I (TM) fuel and a powdered solid waste fuel called ECO-II (TM). Both fuels are sold for $2.00/MBtu FOB Connecticut. This high cost is justifiable as a substitute coal because of its low sulfur content. It is obvious from Figure 11 that if coal-fired equipment is available, this option is economically attractive for smaller military installations.

Figure 12 presents the effect of population size on the operating costs for production of pyrolysis products. In preparing this graph, it was assumed that 80% of the energy in the solid waste was reclaimed in the fuel. However, Garret achieved only

60% conversion for the pyrolytic oil. Therefore, the energy value for No. 6 fuel oil (currently $2.10/MBtu at NWC) must be reduced by a factor of 0.75. Point A on the graph indicates the break-even population required (assuming no credit for dumping fee) for using the pyrolytic oil as a substitute for No. 6 fuel oil (at current prices to NWC). Points B and C likewise correspond to the populations necessary to economically substitute high and low Btu gas, respectively, for natural gas, again using current NWC prices for non-interruptible gas ($0.98/MBtu) and interruptible gas ($0.69/MBtu).

The capital investments required for each of the processes discussed above are plotted against population in Figure 13.

LABORATORY SCALE PYROLYSIS UNIT

A pyrolysis system was designed and fabricated to process a nominal 10 pounds/hour of organic feed in a continuous mode at atmospheric pressure. A diagram of the system is shown in Figure 14, and a photograph of the system is presented in Figure 15. The upper limit on pyrolysis temperature is about 950°C (about 1,750°F), depending upon mass flow rates through the reactor. The stainless steel vessel internal diameter is 3.75 inches and its internal length is 29 inches (0.2 ft^3). The head is secured with eight 5/16-inch bolts and is sealed with a copper gasket.

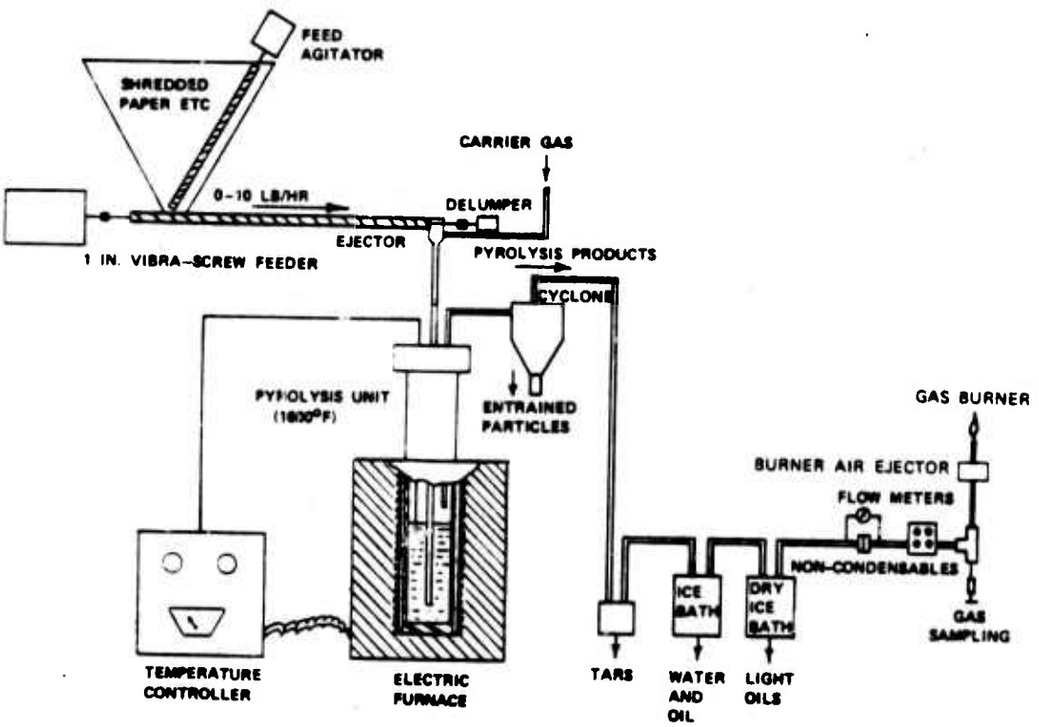

FIGURE 14. Pyrolysis Schematic.

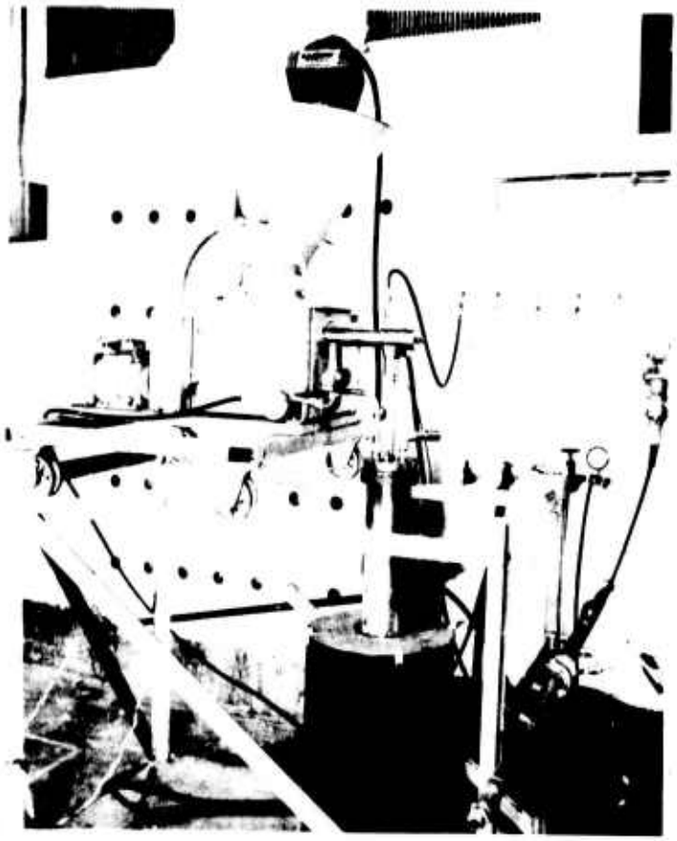

FIGURE 15. Photograph of Lab-Scale Pyrolysis Unit for Solid Waste Conversion.

The solids feeding system is built around a 1-inch Vibra Screw unit. To ensure continuous feeding of shredded organic material, a rotating auger with tines is employed to force the material into the horizontal 1-inch auger. To ensure that the material will fall off the auger into the feeder head, a small bar was attached to the auger shaft. This system has fed up to 18 pounds/hour of 1/4-inch shredded paper. A set of four thin blades revolve at high speed to delump the material as it falls from the auger. The solids are then pumped into the reactor by the ejector action of the pressurized carrier gas. The carrier gas may be easily changed from the control room as desired. The solids are transported to the bottom of the reactor through a 1/2-inch stainless steel pipe. A significant amount of heat transfer into the feed will occur within this pipe. The solids then emerge into the heat transfer medium which may be fluidized particles of char or "sand", or molten salts. The pyrolysis gases and entrained particles of ash, char, and/or fluidizing medium leave the top of the reactor and pass through a short section of stainless steel pipe to a 15-inch diameter stainless steel cyclone separator having a volume of approximately 1 ft^3. The solids are retained in a

receiving vessel at the bottom of the cyclone. After spending several seconds in the cyclone, the gases emerge at the top and pass through a series of air, ice water, and dry ice cooled condensers. Liquid condensate is collected at each of these condensers.

The non-condensible gases then pass through an orifice meter providing instantaneous qualitative flow rates and on through to a Rockwell positive-displacement fuel gas meter which is used to provide true volume flow rate and total volume flow. The orifice and gas meters are used together so that the average molecular weight and mass flow rate of the gases may be determined during the run by the use of a simple nomograph, even though the composition of the gases is unknown.

An air ejector is used to pump the non-condensible gases. This helps to minimize the carrier gas flow rate. The partially pre-mixed gases are then flared-off to prevent any possible hazardous accumulation of flammable and toxic pyrolysis gases. The air ejector and burner will be removed when the synthesis module is added to the pyrolysis module.

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